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7.12.00

DOCKET NO. CH-1999-0004US1

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Box Patent Application
Commissioner of Patents and Trademarks
Washington, D.C. 20231

PATENT FILING TRANSMITTAL

Transmitted herewith for filing is the Patent Application of: Wanda Andreoni and Alessandro Curioni

For: MATERIAL FOR USE IN A LIGHT-EMITTING DEVICE AND HIGHLY EFFICIENT ELECTROLUMINESCENT DEVICE

TYPE OF FILING

This new patent application is for a(n):

[X] Utility
[ ] Design
[ ] Plant
[ ] Divisional
[ ] Continuation
[ ] Continuation-in-part

Benefit of a prior filed application

[ ] This application claims the benefit of an earlier filed U.S. Patent Application under 35 USC 120.
[ ] Please accord Applicant the benefit of the priority date of _____to this case pursuant to 35 USC 119. Applicant's claim for priority is based on application _____filed in _____on that date.

Filing under 37 CFR 1.53 (Utility) or 37 CFR 1.153 (Design)

[X] This is an application filed pursuant to 37 CFR 1.53 or 37 CFR 1.153, permitting receipt of a filing date upon filing of a specification, at least one claim and necessary drawings.
[X] In the event any parts of this application are incomplete, please treat this as a filing under 37 CFR 1.53 or 37 CFR 1.153.

ENCLOSURES

[X] 9 - pages of written description;
[X] 7 - pages of claims;
[X] 1 - pages of abstract;
[ ] 1 - sheets of formal drawings;
[X] ___ - sheets of informal drawings;
[X] Declaration and Power of Attorney or listing of inventors;
and
[X] Two postcards for return to us as proof of receipt of the above documents.

plus
[X] An Assignment of the invention to IBM Corporation and an Assignment cover sheet;
[ ] Verified Statement Claiming Small Entity Status (37 CFR 1.9(f) and 1.27(b))

[ ] Form PTO-1449 (IDS) and two copies of the references listed thereon;
[X] A certified copy of EP99113398.4 (country) patent application number (priority document).
[ ] A preliminary amendment;
[ ] Declaration of Biological Deposit;
[ ] Submission of sequence listing, computer readable copy and/or amendment relating thereto for biotechnology invention containing nucleotide and/or amino acid sequence;
[ ] An associate power of attorney;
[X] Other - Claim for Priority Under 35 U.S.C. § 119

DECLARATION OR OATH

The enclosed Declaration or Oath has been executed by:
[X] Inventor(s);
[ ] Legal representative of the inventors (37 CFR 1.42 or 1.43);
[ ] Joint inventor or person showing proprietary interest on behalf of an inventor who refused to sign or who cannot be reached and this is a petition required by 37 CFR 1.47 and the statement required by 37 CFR 1.47 is attached;
[ ] Has not been executed and is enclosed for the purposes of identifying the inventors.

INVENTORSHIP STATEMENT

The inventorship for all the claims in this application is:
[X] the same;
[ ] not the same and, as an explanation, a statement is/ will be submitted.

LANGUAGE

The application submitted herewith is:
[X] in English;
[ ] in not in English and in terms of 37 CFR 1.52(d) a verified translation is
  [ ] attached
  [ ] not attached.

FEE CALCULATION

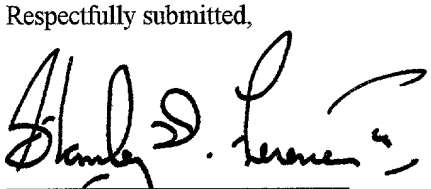
The filing fee has been calculated as shown below:

				OTHER THAN A	
		SMALL ENTITY	OR	SMALL ENTITY	
BASIC FEE Design Patent		*$155*	$	$310	$
BASIC FEE Utility Patent		$345	$	$690	$690
EXTRA FEES		RATE	FEE	RATE	FEE
TOTAL CLAIMS 22 MINUS 20=	2	x 9=	$0	x18=	$36
INDEP.CLAIMS 4 MINUS 3 =	1	x 39=	$0	x78=	$78
[ ] MULTIPLE DEP.CLAIM		+135=	$	+270=	$
[X] ASSIGNMENT		+ 40=	$	+40=	$40
[ ] RULE 53 SURCHARGE		+ 65=	$	+130=	$
TOTAL			$		$844

FEE PAYMENT

[X] Attached is Check No. 5616 in the sum of $ 844.00 to cover the filing fee and, if applicable, the assignment fee.

Respectfully submitted,



Dated: June 11, 2000

Stanley D. Ference III
Reg. No. 33,879

FERENCE & ASSOCIATES
129 Oakhurst Road
Pittsburgh, Pennsylvania 15215
(412) 781-7386
(412) 781-8390-Facsimile

07-12-00

A

PATENT

Docket No. CH9-1999-0004US1

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s)	: Wanda ANDREONI et al	Group Art: not yet assigned
Serial No.	: not yet assigned	Examiner: not yet assigned
Filed	: herewith	
For	: MATERIAL FOR USE IN A LIGHT-EMITTING DEVICE AND HIGHLY EFFICIENT ELECTROLUMINESCENT DEVICE	

EXPRESS MAIL CERTIFICATE

Express Mail Label No. EL503717332US

Date of Deposit 11 June 2000

I hereby certify that the following attached paper(s) or fee:

Patent Application
- Written Description
- Claims 1-22
- Abstract
- Drawings (Fig. 1)

Declaration and Power of Attorney
Assignment with Completed Cover Sheet
Claim for Priority
Certified Copy of Priority Document
Patent Filing Transmittal
Certificate of Express Mail
Check in the Amount of $844 (Check No. 5616)
Two Return Postcards

are being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 C.F.R. 1.10 on the date indicated above and is addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231.

Stanley D. Ference III
(Typed or printed name of person mailing paper)

(Signature of person mailing paper(s) or fee)

Mailing Address:

FERENCE & ASSOCIATES
129 Oakhurst Road
Pittsburgh, Pennsylvania 15215
(412) 781-7386
(412) 781-8390-Facsimile

MATERIAL FOR USE IN A LIGHT-EMITTING DEVICE AND HIGHLY EFFICIENT ELECTROLUMINESCENT DEVICE

Field of the Invention

The present invention relates generally to a material for use in a light-emitting device, and more particularly to increasing the efficiency of organic light-emitting devices (OLEDs).

Background of the Invention

Electroluminescent devices based on organic thin layers are light-emitting devices similar to semiconductor-based light-emitting diodes, and are currently being considered for the next generation of flat panel displays. Structurally, these devices contain spaced electrodes separated by an electroluminescent medium which emits light in response to the application of an electrical potential difference across the electrodes.

Preferred forms of OLEDs typically include an anode, an organic hole injecting and transporting zone in contact with the anode, an electron injecting and transporting zone forming a junction with the organic hole injecting and transporting zone, and a

cathode in contact with the electron injecting and transporting zone. By application of an electric potential across the electrodes, holes and electrons are injected into the organic zones from the anode and cathode, respectively. Light emission results from the hole-electron recombination within the subject device. This carrier recombination generates excited molecules, which eventually emit light or become thermally deactivated. The efficiency of present OLEDs is thus highly dependent upon both carrier recombination efficiency and photoluminescence quantum yield of the emitting material.

In OLEDs based on small molecules, one of the common electroluminescent units is a highly flourescent aluminum complex, tris(8-quinolinolato)aluminum(III) (Alq3). This flourescent aluminum complex (Alq3) emits a green light and fulfills a number of prerequisites, including but not limited to, when used the device is stable and is quite suitable for carrier transport. The luminescence yield, however, is relatively low. Attempts to improve the luminescence yield have been made by doping the Alq3 layer with fluorescent dye molecules.

Kido et al., Appl. Phys. Lett., vol. 73, no. 19, pages 2721-2723, dated November 9, 1998, discloses an efficient organic electroluminescent device which is fabricated by using tris(4-methyl-8-quinolinolato)aluminum(III) (Almq3) as an emitter layer.

Additionally, to using this complex, a multi-layer device structure consisting of a hole-injecting layer, a hole transport layer, a dye-doped Almq3 emitting layer, and an electron transport layer was employed in order to reduce the driving voltage as well as to maximize carrier recombination efficiency. Kido et al. reports a maximum luminescence of over 140 000 cd/m^2 and an external quantum efficiency of 7.1%, which is believed to be the highest efficiency ever reported for organic devices.

US-A-5,150,006, discloses an internal junction organic electroluminescent device comprised of, in sequence, an anode, an organic hole injecting and transporting zone, an organic electron injecting and transporting zone, and a cathode. The organic electron injecting and transporting zone is comprised of an electron injecting layer in contact with the cathode. Interposed between the electron injecting layer and the organic hole injecting and transporting zone is a blue emitting luminescent layer comprised of an aluminum chelate containing a phenolato ligand and two R^s-8-quinolinolato ligands, where R^s substituents are used to block the attachment of more than two substituted 8-quinolinolato ligands to the aluminum atom. The presence of the phenolato ligand shifts the device emission to the shorter blue wavelengths of the spectrum and increases emission efficiency. Increased operating stability can be realized by the incorporation of a pentacarboxyclic aromatic fluroescent dye.

US-A-5,456,988 discloses an electroluminescent (EL) device including an organic electron transport layer comprising Alq3 substituted with Cl or Br in the 5-position. A useful EL device is provided that has excellent durability and retains stable luminescence for a long period of time by using a compound other than the 8-quinolinolato-aluminum complex as an emitting material. No values of luminescence for the single halogen substitution, however, are reported.

A need has thus been recognized to enhance the luminescence of OLEDs.

Summary of the Invention

The present invention, in accordance with at least one presently preferred embodiment, exploits an organic material having tris(8-quinolato)aluminum(III) (Alq3) as a base unit. Consequently, the present invention broadly contemplates use of an organic material in light-emitting devices to increase intrinsic luminescence of the organic molecular unit.

In one aspect of the present invention, there is provided a Alq3-based material in which the intrinsic luminescence of the organic molecular unit is directly enhanced by a modification to the relevant electron states, preferably through specific substitutions on the quinolate ring, namely by substituting the Alq3 unit in positions 3 or 4 and 5. An electron-donor group is substituted in the 3- or 4-position and an electron-acceptor or p-delocalizing group is substituted in the 5-position, preferably simultaneously. The combined substitution amplifies the enhancement of the luminescence and reduces the induced shift of the ionization potential and electronic affinity values with respect to single substitutions which is important to incorporate the new compounds in available device structures.

In another aspect of the present invention, an electroluminescent device is provided which comprises an anode, an organic hole injecting and transporting zone, an organic electron injecting and transporting zone, a cathode and a luminescent layer of tris(8-quinolinolato)aluminum(III) (Alq3), wherein said Alq3 is substituted in the 3- or 4-position with an electron-donor group and simultaneously substituted in said 5-position with an electron-acceptor or a p-delocalizing group.

For a better understanding of the present invention, together with other and further features and advantages thereof, reference is made to the following description, taken in conjunction with the accompanying drawings, and the scope of the invention will be pointed out in the appended claims.

Brief Description of the Drawings

Figure 1 illustrates the formula of an Alq3 where the atoms on the quinolinolato ligand are labeled with standard notation.

Detailed Description of the Preferred Embodiments

Referring now to Figure 1, the formula of Alq3 is shown and the location of the atoms on the quinolinolato ligand are labeled with the standard notation. The relatively weak (compared with other fluorescent organic materials) luminescence of the Alq3 molecule is associated with different spatial localization of the electron states involved in the luminescence process which limits the corresponding transition probability. Specifically, the holes acceptor states (Highest Occupied Molecular Orbital (HOMO) set of states) are localized mainly in the phenoxyde side of the ligands whereas the electron

acceptor states (Lowest Unoccupied Molecular Orbital (LUMO) set of states) are localized mainly on the pyridyl side of the ligands. HOMO-LUMO electronic transitions (the relevant ones for luminescence properties) are thus limited.

An increase in the intrinsic luminescence yield can be achieved by modifying the relevant electron states by means of specific chemical substitutions on the quinolate rings which improve the spatial overlap between the HOMO and LUMO set of states, and therefore, indirectly the transition probability and hence the intrinsic luminescence yield. In accordance with the present invention, substitutions are made using an electron-donor group (R^{py}) in the 3- or 4-position and, at the same time, an electron-acceptor or p-delocalizing group (R^{ph}) in the 5-position.

The substituents R^{py} are preferably selected from the groups CR'R"R'", -NR2 and -O-R, wherein R, R', and R" = (H, Alkyl), R'" = (Alkyl), and may be generally selected from any group that is able "to push" electrons onto the ligands. The substituents R^{ph} are preferably selected from the groups -CX3, wherein X=F, Cl, Br, -CX2-CX3, -SO3-R, CR=CR2, -CX=CX2, -COOR, -SO3M, and -COOM, wherein M = metal ion, R = alkyl and X = F, Cl, Br, and may be generally selected from any group that is able to strongly

draw electrons from the ligand or enhance the p-conjugation (delocalization) on the phenoxide-side of the ligand.

An electroluminescent device using the Alq3 derivatives of the present invention preferably consists of a hole injection electrode, an electron injection electrode and at least one organic emitting layer incorporating at least one of the proposed Alq3 derivatives. It should be understood that the electroluminescent device may contain additional hole-transport layers between the hole injection layer and the organic emitting layers and/or additional electron transport layers between the electron injection electrode and the organic light-emitting layers.

It is to be further understood that since the organic light-emitting layer consists of Alq3 derivatives having a larger intrinsic luminescence yield with a calculated enhancement factor up to four, the device will have a larger quantum efficiency than any other device made by unsubstituted and undoped Alq3. Moreover, since this is obtained by directly modifying the Alq3 molecule and without adding any highly fluorescent dopants, all contrary to the work known from the prior art, the good stability and carrier transport properties of the Alq3 layers are preserved, and no additional energy transfer step from Alq3 to the dopant molecules is needed to have high luminescence yield.

If not otherwise stated herein , it is to be assumed that all patents, patent applications, patent publications and other publications mentioned and cited herein are hereby fully incorporated by reference herein as if set forth in their entirety herein.

Although illustrative embodiments of the present invention have been described herein with reference to the accompanying drawing, it is to be understood that the invention is not limited to those precise embodiments, and that the various other changes and modifications may be affected therein by one skilled in the art without departing from the scope or spirit of the invention.

Claims

What is claimed is:

1. A compound as shown in Figure 1, wherein:

a base unit consists of tris (8-quinolinato)aluminum(III) (Alq3);

said base unit in the 3- or 4-position is substituted with an electron-donor group; and

said base unit in the 5-position is simultaneously substituted with an electron-acceptor or p-delocalizing group.

2. The compound according to Claim 1, wherein said electron-donor group in said 3- or 4-position is selected from a group consisting of -CR'R"R'", NR2, and -OR, wherein R, R', R" = H or Alkyl, and R'" = Alkyl.

3. The compound according to Claim 1, wherein said electron-acceptor or p-delocalizing groups in said 5-position are selected from a group consisting of -CX3, -CX2,-CX3, -SO3R, -CR=CR2, -CX=CX2, -COOR, -SO3R, -SO3M and -COOM, whereby X=F, Cl, Br; R = H or Akyl, and M = metal ion.

4. The compound according to Claim 2, wherein said electron-acceptor or p-delocalizing groups in said 5-position are selected from a group consisting of -CX3, -CX2,-CX3, -SO3R, -CR=CR2, -CX=CX2, -COOR, -SO3R, -SO3M and -COOM, whereby X=F, Cl, Br; R = H or Akyl, and M = metal ion.

5. The compound according to Claim 1, wherein said electron-donor group in the said 3- or 4-position is -CH3 and said electron-acceptor group in said 5-position is -CF3.

5. The compound according to Claim 1, wherein said electron-donor group in the said 3- or 4-position is -OR and said electron-acceptor group in said 5-position is -CF=CF2.

7. The compound according to Claim 1, wherein said electron-donor group in the said 3- or 4-position is -CH3 and said electron-acceptor group in said 5-position is -CF=CF2.

8. A organic material having tris (8-quinolinato)aluminum(III) (Alq3) as a base unit and wherein:

said base unit in the 3- or 4-position is substituted with an electron-donor group; and

said base unit in the 5-position is simultaneously substituted with an electron-acceptor or p-delocalizing group.

9. The material according to Claim 8, wherein said electron-donor group in said 3- or 4-position is selected from a group consisting of -CR'R"R'", NR2, and -OR, wherein R, R', R" = H or Alkyl, and R'" = Alkyl.

10. The compound according to Claim 8, wherein said electron-acceptor or p-delocalizing groups in the said 5-position are selected from a group consisting of -CX3, -CX2,-CX3, -SO3R, -CR=CR2, -CX=CX2, -COOR, -SO3R, -SO3M and -COOM, whereby X=F, Cl, Br; R = H or Akyl, and M = metal ion.

11. The material according to Claim 9, wherein said electron-acceptor or p-delocalizing groups in said 5-position are selected from a group consisting of -CX3, -CX2,-CX3, -SO3R, -CR=CR2, -CX=CX2, -COOR, -SO3R, -SO3M and -COOM, whereby X=F, Cl, Br; R = H or Akyl, and M = metal ion.

12. The material according to Claim 8, wherein said electron-donor group in the said 3- or 4-position is -CH3 and said electron-acceptor group in the said 5-position is -CF3.

13. The material according to Claim 8, wherein said electron-donor group in said 3- or 4-position is -OR and said electron-acceptor group in said 5-position is -CF=CF2.

14. The material according to Claim 8, wherein said electron-donor group in the said 3- or 4-position is -CH3 and said electron-acceptor group in said 5-position is -CF=CF2.

15. An electroluminescent device comprising:

an anode,

an organic hole injecting and transporting zone,

an organic electron injecting and transporting zone;

a cathode; and

a luminescent layer of the compound shown in Figure 1, wherein said compound is substituted in the 3- or 4-position with an electron-donor group and simultaneously substituted in said 5-position with an electron-acceptor or a p-delocalizing group.

16. An electroluminescent device according to Claim 15, wherein said electron-donor group in the 3-or 4-positions is selected from the group consisting of -CR'R"R'", NR2, and -OR, wherein R, R', R"=H or Alkyl and R'"=Alkyl.

17. An electroluminescent device according to Claim 15, wherein said electron-donor or p-delocalizing groups in the 5-position are selected from the group consisting of -CX3, -CX2,-CX3, -SO3R, -CR=CR2, -CX=CX2, -COOR, -SO3M, and -COOM, whereby X = F, Cl, Br; R = H or Alkyl and M = metal ion.

18. An electroluminescent device according to Claim 16, wherein said electron-donor or p-delocalizing groups in the 5-position are selected from the group consisting of -CX3, -CX2,-CX3, -SO3R, -CR=CR2, -CX=CX2, -COOR, -SO3M, and -COOM, wherein X = F, Cl, Br; R = H or Alkyl and M = metal ion.

19. An electroluminescent device comprising:

an anode,

an organic hole injecting and transporting zone,

an organic electron injecting and transporting zone;

a cathode; and

a luminescent layer of tris(8-quinolinolato)aluminum(III) (Alq3), wherein said Alq3 is substituted in the 3- or 4-position with an electron-donor group and simultaneously substituted in said 5-position with an electron-acceptor or a p-delocalizing group.

20. An electroluminescent device according to Claim 19, wherein said electron-donor group in said 3-or 4-position is selected from the group consisting of -CR'R"R"', NR2, and -OR, wherein R, R', R" = H or Alkyl and R"' = Alkyl.

21. An electroluminescent device according to Claim 19, wherein said electron-donor or p-delocalizing groups in said 5-position are selected from the group consisting of -CX3, -CX2,-CX3, -SO3R, -CR=CR2, -CX=CX2, -COOR, -SO3M, and -COOM, whereby X = F, Cl, Br; R = H or Alkyl and M = metal ion.

22. An electroluminescent device according to Claim 20, wherein said electron-donor or p-delocalizing groups in said 5-position are selected from the group consisting of -CX3, -CX2,-CX3, -SO3R, -CR=CR2, -CX=CX2, -COOR, -SO3M, and -COOM, whereby X = F, Cl, Br; R = H or Alkyl and M = metal ion.

5

MATERIAL FOR USE IN A LIGHT-EMITTING DEVICE AND HIGHLY EFFICIENT ELECTROLUMINESCENT DEVICE

Abstract of the Disclosure

A material is provided that can be used for a light-emitting device. The base unit of said material is tris(8-quinolinolato)aluminum(III) (Alq3). This Alq3 is substituted in the said 3- or 4-position with an electron-donor group and simultaneously in the said 5-position with an electro-acceptor or p-delocalizing group. Using this material as an emitting luminescent layer, the efficiency of the intrinsic luminescence can be greatly enhanced.

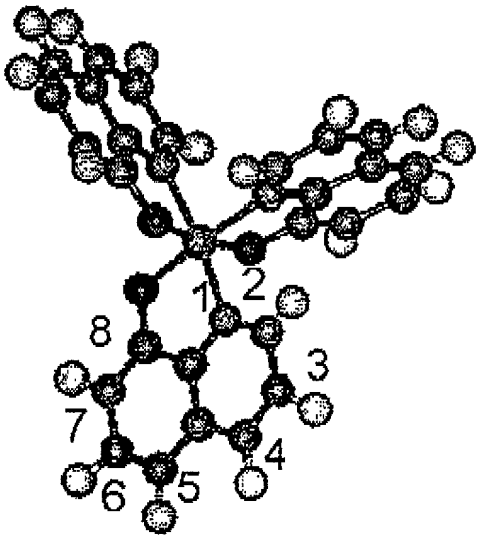


FIG. 1

DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name;

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

MATERIAL FOR USE IN A LIGHT-EMITTING DEVICE AND HIGHLY EFFICIENT ELECTROLUMINESCENT DEVICE

the specification of which (check one)

__X__ is attached hereto.

__X__ was filed on _____ as International Business Machines Docket No. CH9-1999-0004US1

and was amended on _____ (if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application, having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

				Priority Claimed
99113398.4	EP	12/07/1999		__X__ Yes ___ No
(Number)	(Country)	(Day/Month/Year Filed)		

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

(Application Number)	(Filing Date)
(Application Number)	(Filing Date)

DELARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION

I hereby claim the benefit under 35 U.S.C. §120 of any United States Application(s), or §365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States, or PCT International application in the manner provided by the first paragraph of 35 U.S.C. §112, I acknowledge the duty to disclose information material to the patentability of this application as defined in 37 CFR §1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith (list name and registration number).

Manny W. Schecter (Reg. 31,722), Terry J. Ilardi (Reg. 29,936), Christopher A. Hughes (Reg. 26,914), Edward A. Pennington (Reg. 32,588), John E. Hoel (Reg. 26,279), Joseph C. Redmond, Jr. (Reg. 18,753), Paul J. Otterstedt (Reg. 37,411), Douglas W. Cameron (Reg. 31,596), Wayne L. Ellenbogen (Reg. No. 43,602), Stephen C. Kaufman (Reg. 29,551), Daniel P. Morris (Reg. 32,053), Louis J. Percello (Reg. 33,206), Jay P. Sbrollini (Reg. 36,266), Robert M. Trepp (Reg. 25,933), David M. Shofi (Reg. 39,835, and Louis P. Herzberg (Reg. 41,500)

Send Correspondence to: FERENCE & ASSOCIATES, 129 Oakhurst Road, Pittsburgh, PA 15215

Direct Telephone Calls to: (name and telephone number) Stanley D. Ference III, (412) 781-7386

DELARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION

Wanda Andreoni
Full name of sole or first inventor

W Andreoni — July 5, 2000
Inventor's Signature — Date

Obstgartenstrasse 22, CH-8136 Gattikon, Switzerland
Residence

Switzerland
Citizenship

Same as above
Post Office Address

Alessandro Curioni
Full name of second joint-inventor, if any

A Curioni — July 5, 2000
Inventor's Signature — Date

Obstgartenstrasse 28, CH-8136, Gattikon, Switzerland
Residence

ITALY
Citizenship

Same as above
Post Office Address